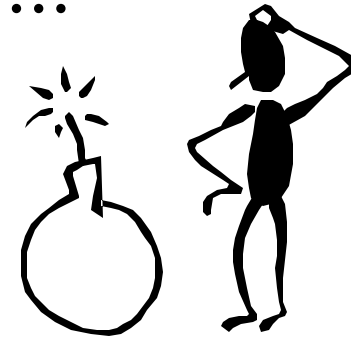# Feasible-Sets: 1D and the general phase problem

Subtitle:

If the solution is not unique…

should we go fishing?

L. D. Marks

Department of Materials Science & Engineering

Northwestern University

Overview & Acknowledgements

- Feasible-set approach to crystallographic problems

  Eric Landree, Wharton Sinkler & **Erman Bengu ++**

- 1D Josephson junction problem

  Mike Carmody & Karl Merkle

- 1D x-ray reflectivity problem

  Erman Bengu & **Monica Salud**

- 1D/3D Surface Phase Problem

  Collaborators (data): **Robert Feidenhans'l**, Joerg Zegenhagen, Antoine Barbier,….
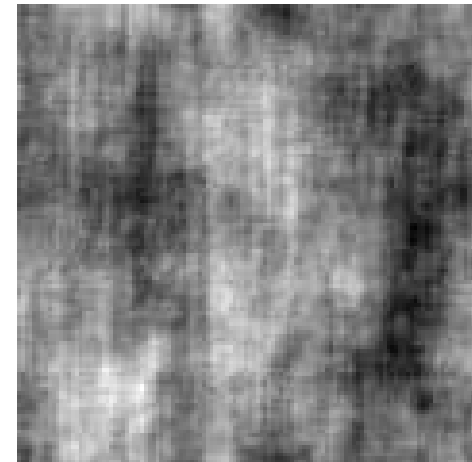
Basics

- We know the amplitudes
- We want to find the phases
- Problem is insolvable without additional information – constraints
- Use an iterative approach

The importance of phase information

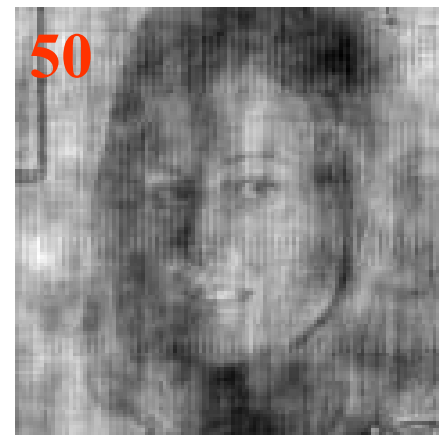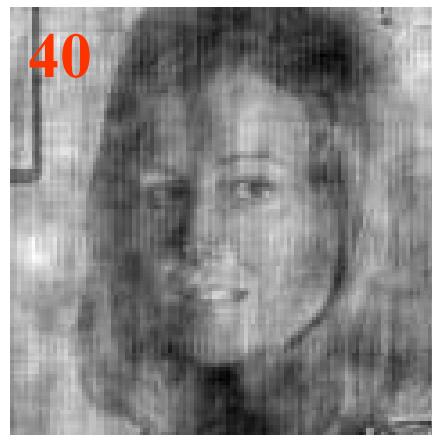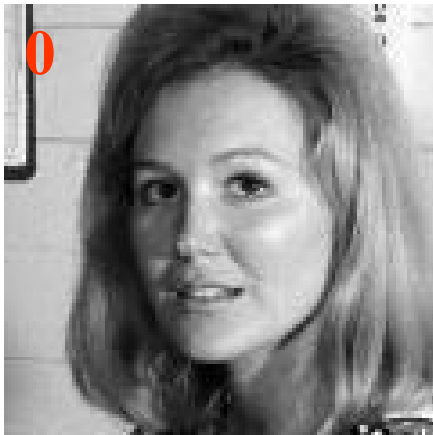Correct Modulus
Random Phases
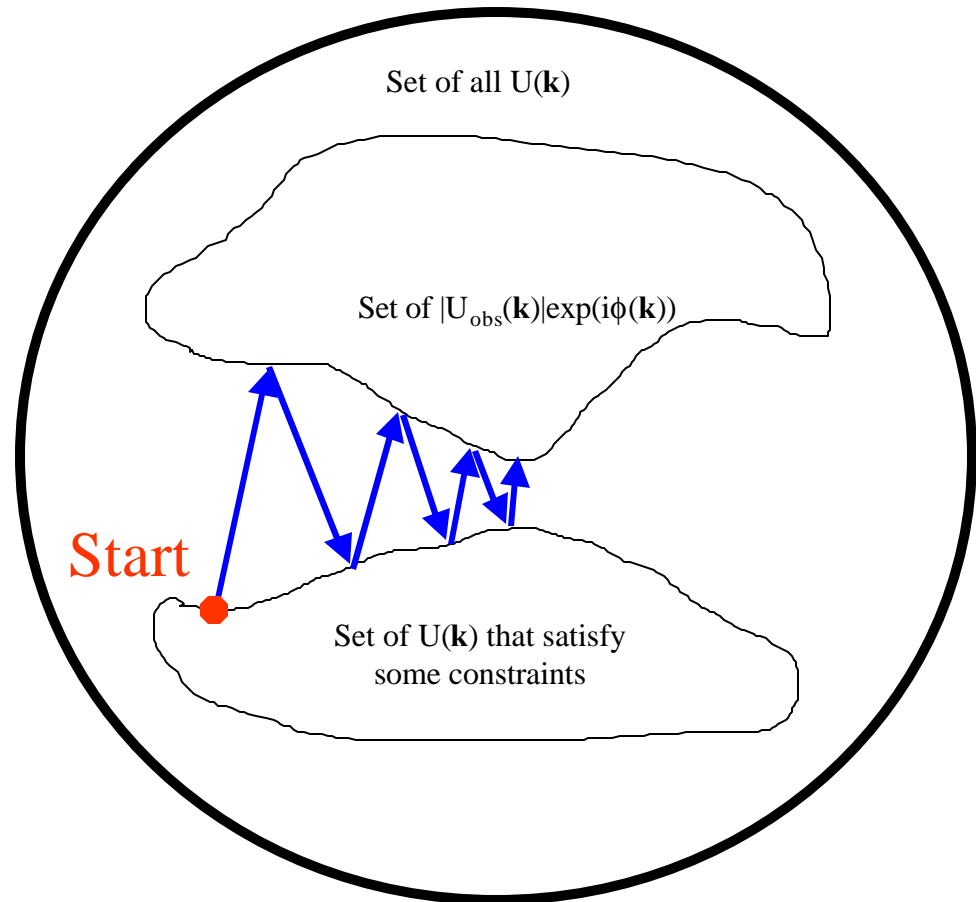
Suzy

Correct Phase
Random Modulus

Role of error in phases (degrees)

We would like to find the phases exactly, but we don't have to

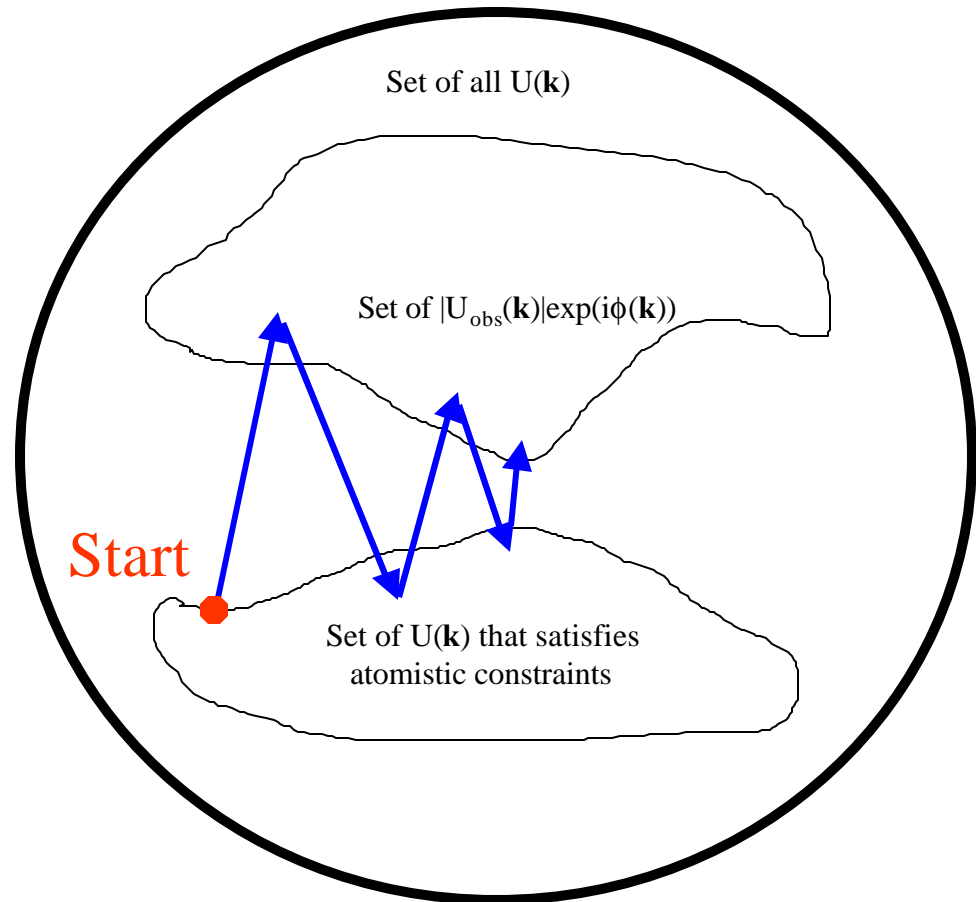# Algorithm Overview (Gerschberg-Saxton)

Successive Projections

- Iterate between projections
- Other variants possible (see Combettes, *Advances in Imaging and Electron Physics* **95,** 155-270, 1996)
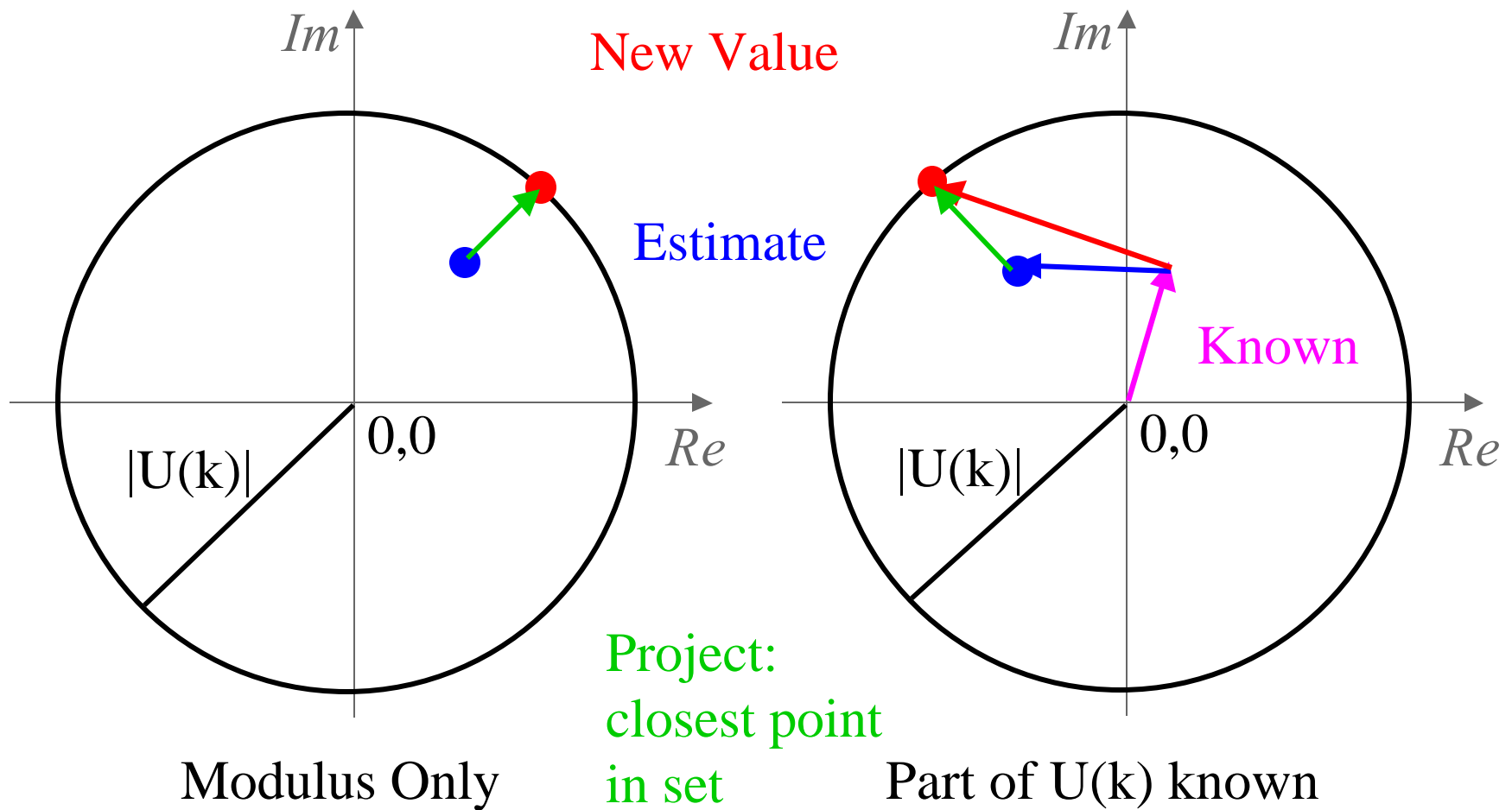
Set of all U($\mathbf{k}$)

Set of |U$_{obs}$($\mathbf{k}$)|exp(iϕ($\mathbf{k}$))

Start

Set of U($\mathbf{k}$) that satisfy some constraints

Over-relaxed Projections

- Iterate between projections

- Overshoot (deliberately)

- Converges faster

- Sometimes better solutions

Orthogonal Projections

New Value

Estimate

Im

Re

0,0

$|U(k)|$

Modulus Only

Im

Re

0,0

$|U(k)|$

Known

Project:
closest point
in set

Part of U(k) known

Example: Fourier Difference Map

- We know all the moduli, $|U(k)|$
- Suppose we know part of the structure,

  $U_a(k) = |U_a(k)|\exp(i\phi_a(k))$

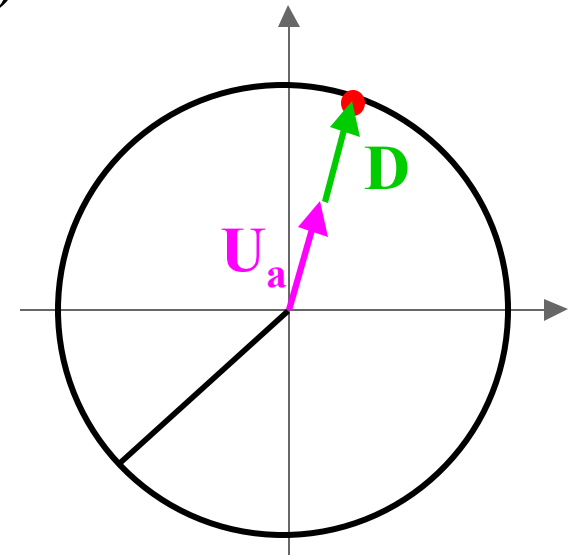- Find the additional component $D(k)$ such that

  $|U_a(k) + D(k)| = |U(k)|$

- Minimize (orthogonal projection):

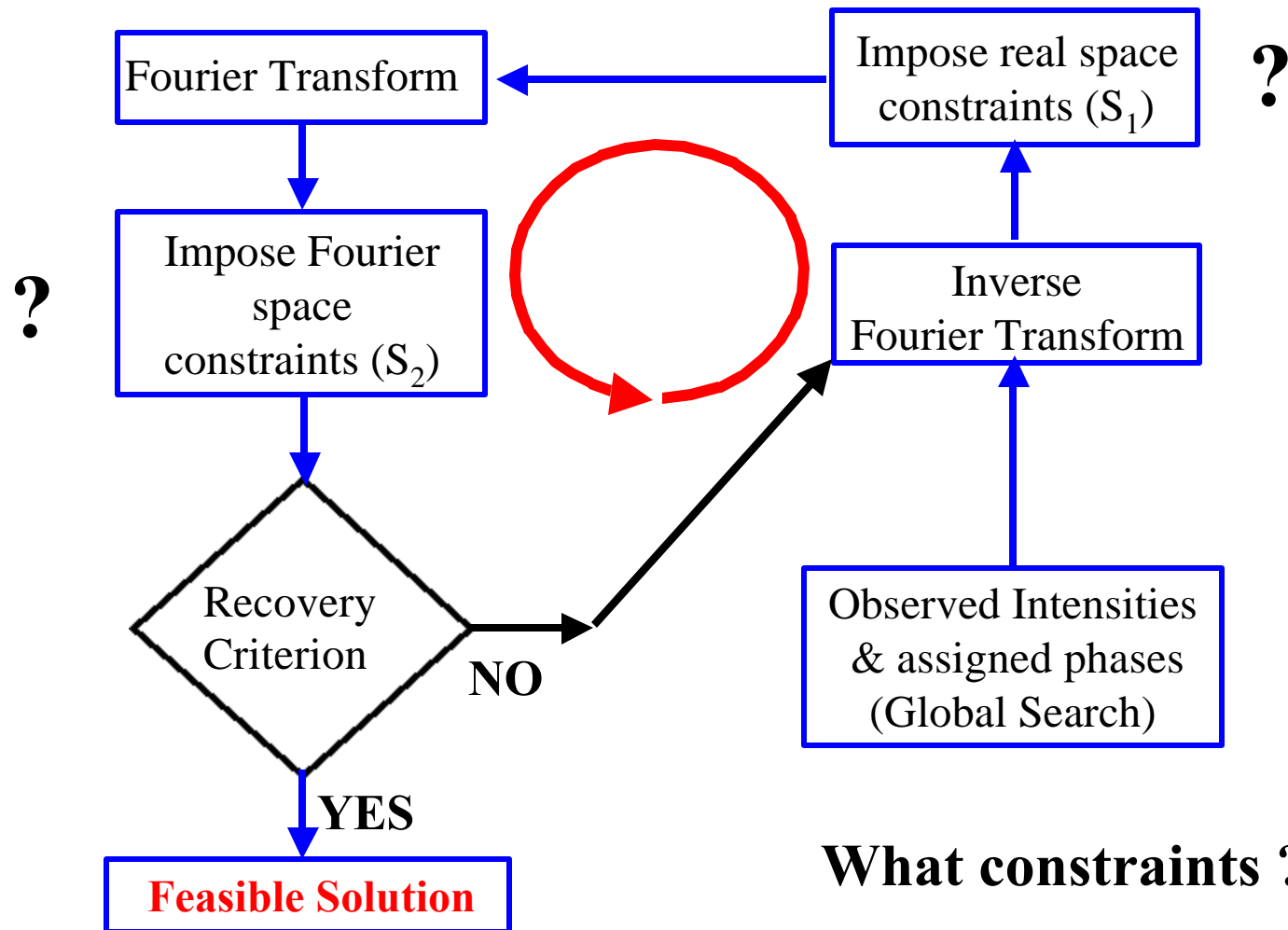 $|D(k)|^2 - \lambda\{\,|U_a(k)+D(k)|\text{-}|U(k)|\}$

- Solution

  $D(k) = \exp(i\phi_a(k))\{|U(k)|\text{-}|U_a(k)|\}$

  Conventional Fourier Difference Map

# Algorithm Overview (Gerschberg-Saxton)

Where do constraints come from

- **Physical nature of experiment**
  - Limited beam or object size
- **Physical nature of scattering**
  - Atomic scattering
- **Statistics & Probability**
  - Minimum Information/Bias = Maximum Entropy

The $64,000 question

- Consider the points which obey a constraint as a set
- A set is convex if any point between two members is also a member
- Amplitude measurements do not form a convex set

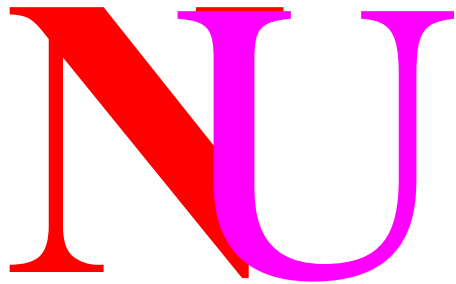$|F(k)|$=const

A

C

B
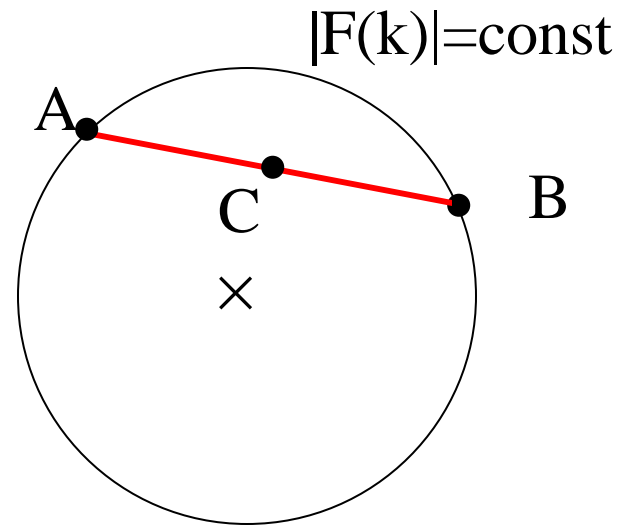
Types of Constraints

- **Convex – highly convergent**
  - Multiple convex constraints are unique
- **Non-convex – weakly convergent**
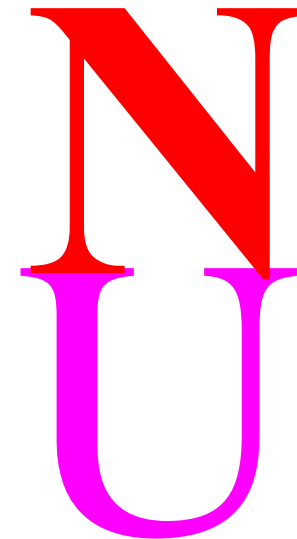 - Multiple non-convex constraints may not be unique

Multiple non-convex constraints

Consider the two sets "N" and "U"

Overall Convex

Overall Non-Convex

Simplest Constraint: Limited Object

- 1D Continuous, overall problem is non-unique

- 1D Non-continuous, may be unique

- nD Continuous, $n \geq 2$, overall unique

(Provided that the Patterson Function is limited)
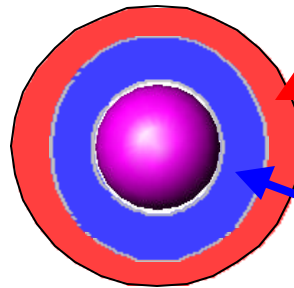
Other Constraints

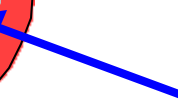| Convex	Non-Convex
Positivity (weak)	Presence of Atoms
Atoms at given positions	Bond Lengths
Least bias (MaxEnt)	Interference $A(k)=\mid B(k)+Known(k)\mid^2$
Intensities & errors $\equiv \chi^2$	Anti-bumping
Statistics (e.g. Σ_2)	Bond angles
Support for gradient	
Symmetry	

Atomistic Constraints

$\rho(r)$ known (convex if position is known)

Bonding – another atom

Bumping $\rho(r)=0$

Atomistic Constraint

- **Simple case, Unitary Sayre Equation**
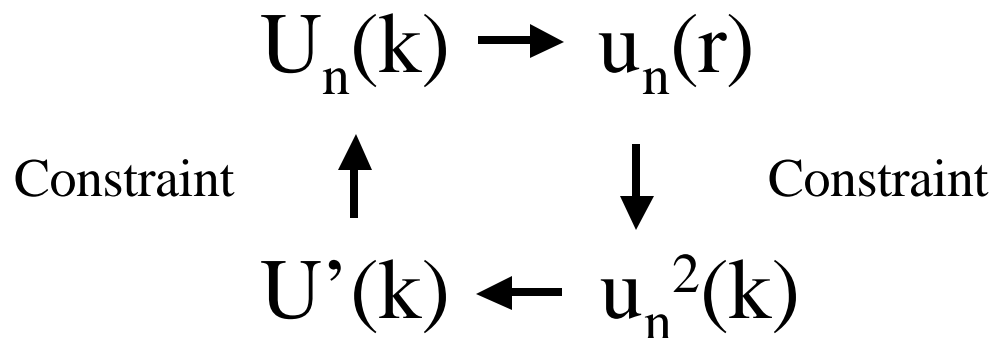 - $F(k) = \sum_l f(k)\exp(2\pi i k.r_l)$
- **Divide by N, #atoms & f(k), atomic scattering factors**
  - $U(k) = (1/N)\sum_l \exp(2\pi i k.r_l)$; $u(r) = (1/N)\sum_l \delta(r-r_l)$
  - $u(r) = Nu(r)^2$

Classic Direct Methods

- ■ Consider as an iteration

$$U_n(k) \longrightarrow u_n(r)$$

Constraint $\uparrow$ $\downarrow$ Constraint

$$U'(k) \longleftarrow u_n^2(k)$$

- ■ Note the similarities

  – Tangent Formula $\equiv$ Orthogonal Projection

  – Real space operator, effectively an eigenfunction (fixed point) method
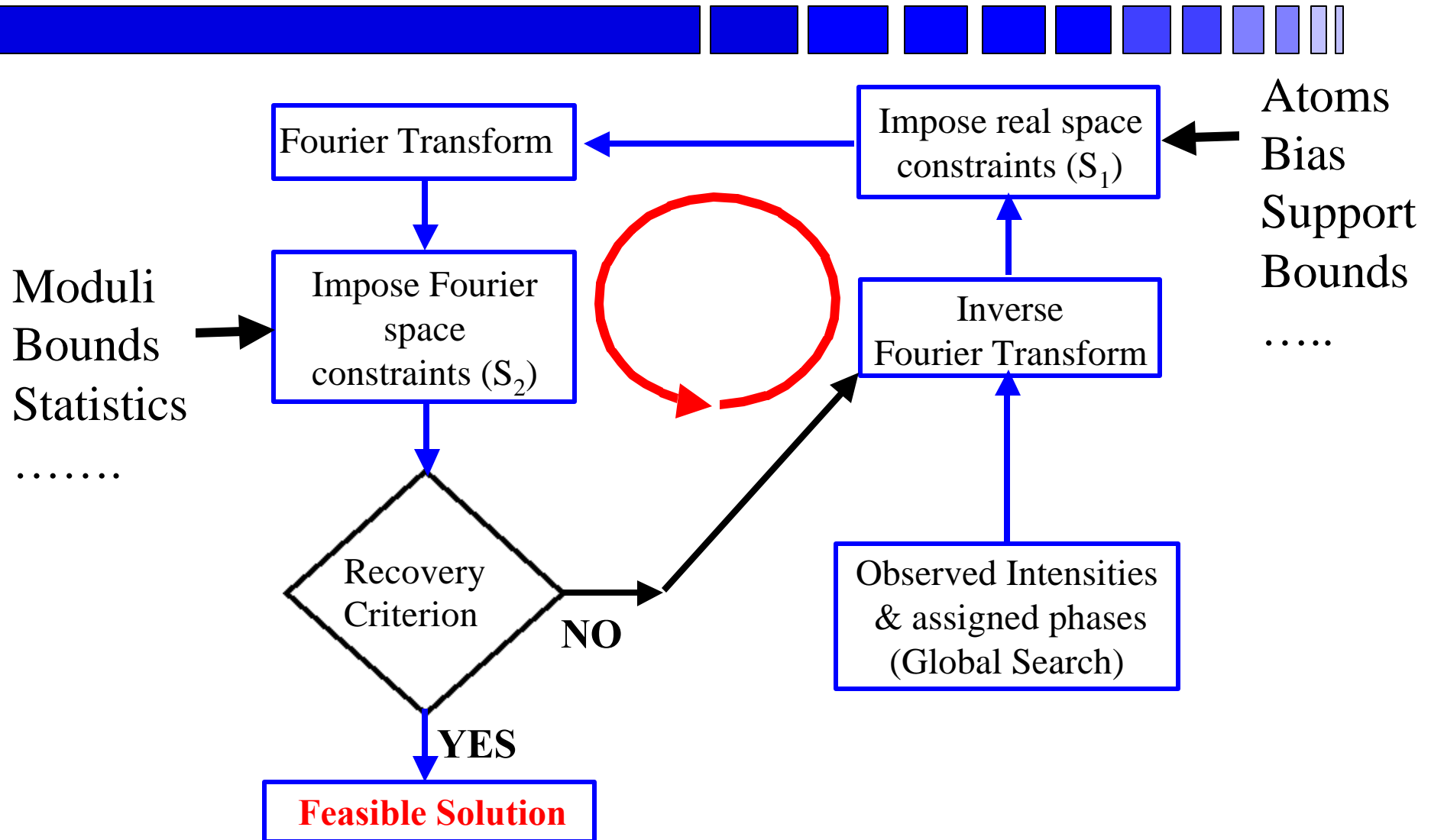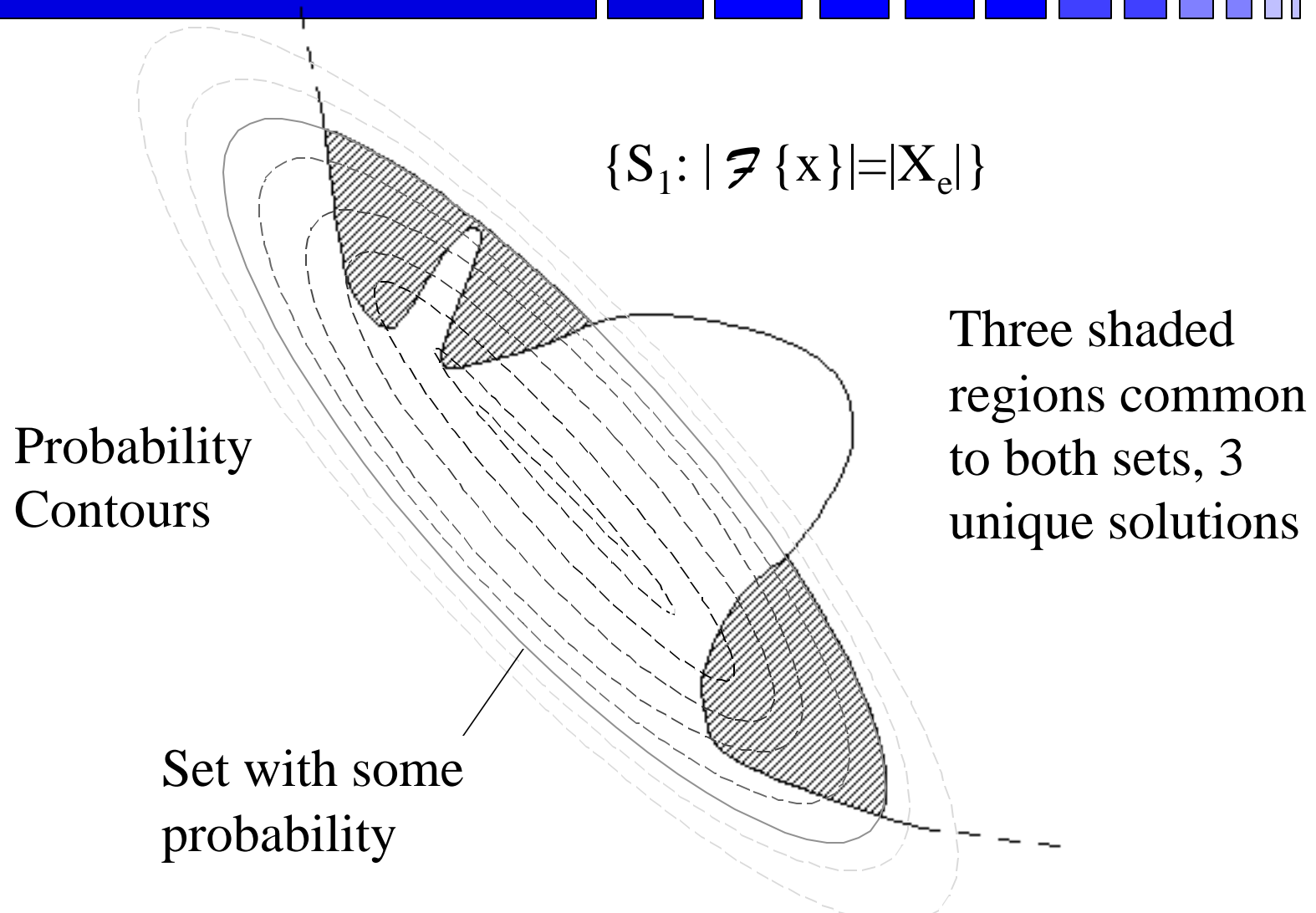
Null Hypothesis: Minimum Bias/Information

- **Consider most probable distribution of phases for random atoms**
  - Central-Limit Theorem
    - Cochran Distribution
  - Bregman Projection using xlogx
    - Maximum entropy or Kullback-Leibler metric

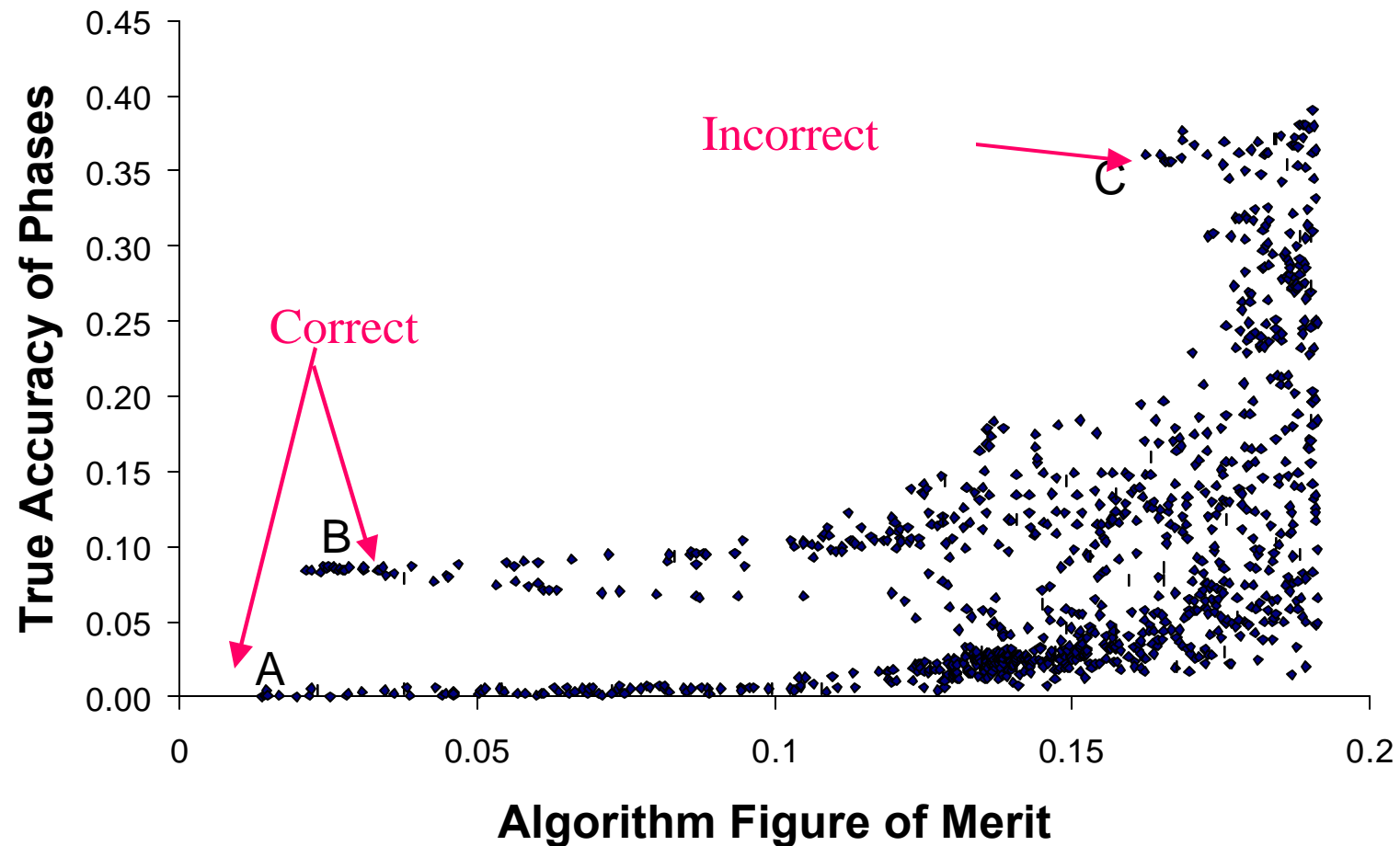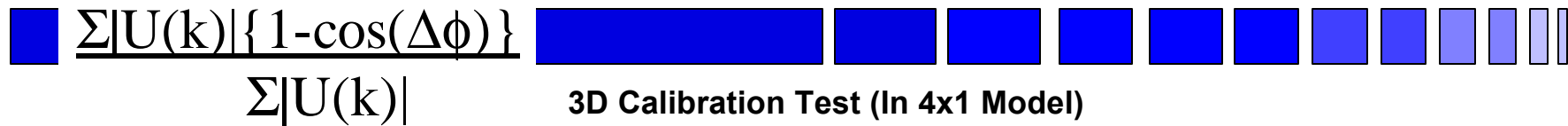Convex constraints

Algorithm Overview (Gerschberg-Saxton)

Fourier Transform

Impose real space constraints (S_1)

Atoms
Bias
Support
Bounds
.....

Moduli
Bounds
Statistics
....

Impose Fourier space constraints (S_2)

Inverse Fourier Transform

Recovery Criterion

Observed Intensities & assigned phases (Global Search)

NO

YES

Feasible Solution

Multiply-Connected Feasible Set

$\{S_1: |\mathcal{F}\{x\}|=|X_e|\}$

Probability
Contours

Three shaded
regions common
to both sets, 3
unique solutions

Set with some
probability

Typical results

$\Delta\phi$ = phase error

$$\frac{\Sigma|U(k)|\{1-\cos(\Delta\phi)\}}{\Sigma|U(k)|}$$

3D Calibration Test (In 4x1 Model)

Hypothesis (5/18/2001)

- Think about the probability near a solution
- Apply classic D.M. statistics to
  $$U_{n+1}(k)-U_n(k)$$
- Use central-limit theorem
  - $P(|U(k)|\cos(\Delta\phi))\sim C\exp\{N|U(k)|^2\cos(\Delta\phi)\}$
  - $\Delta\phi$ = phase error
- Needs verification – but correllates with results!

Crystallographic methodology

Overall Unique

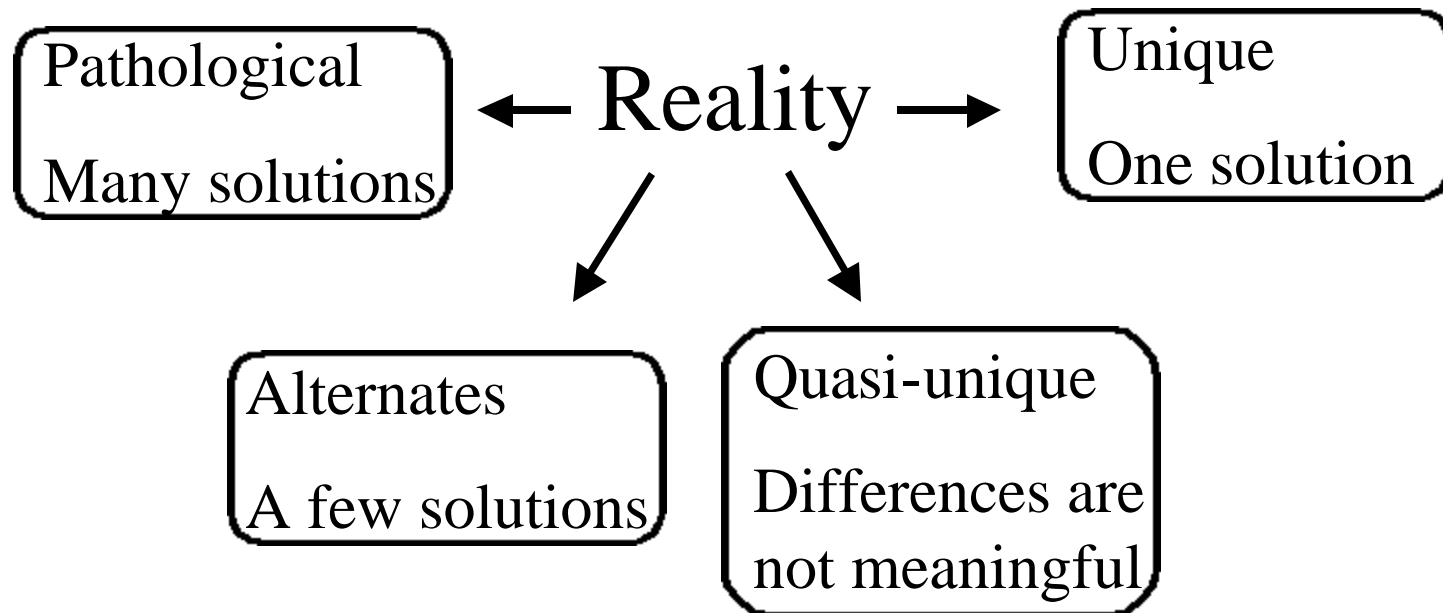Addition of additional convex constraints tends to give a unique solution

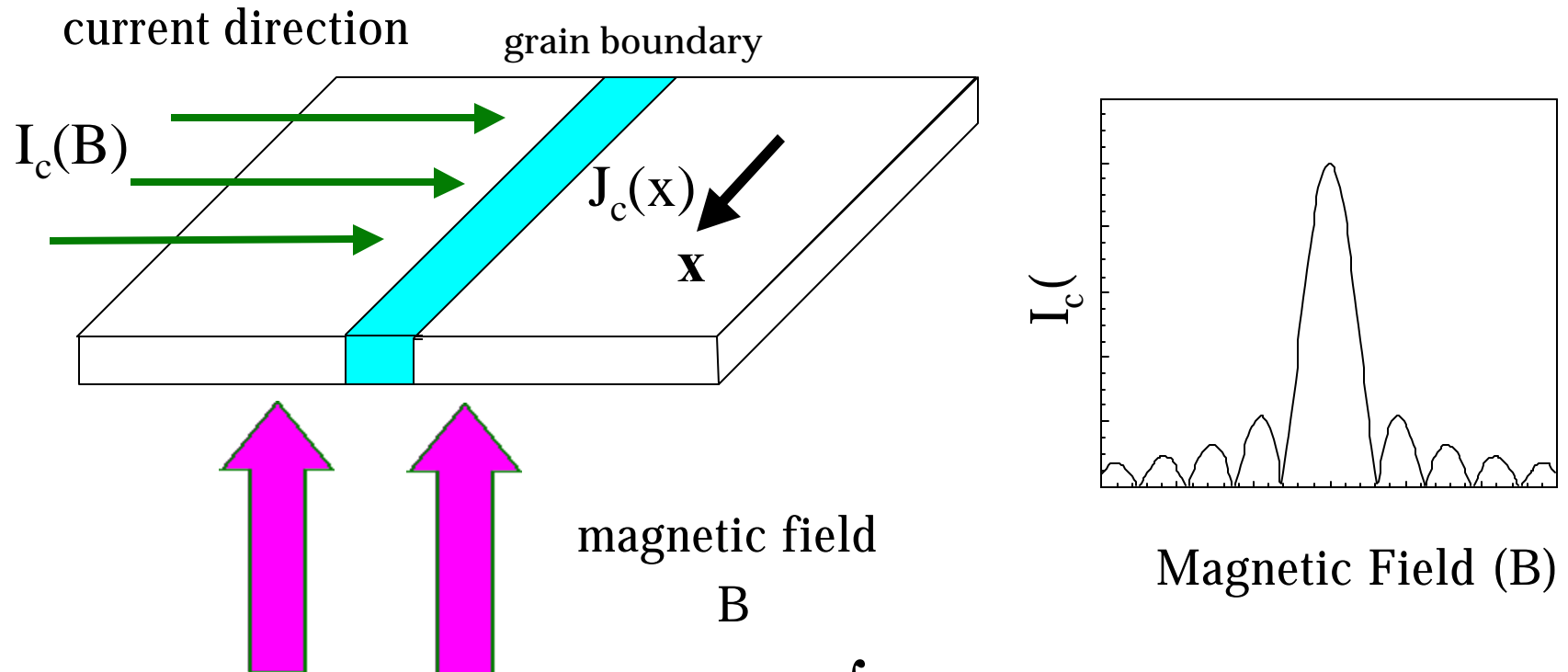Structure Completion: add additional constraints as the phases become known

N U O

Overall Non-Convex

1D Support Constraint

- "Conventional Wisdom"
  - In 1D, overall problem is non-unique

Pathological

Many solutions ← Reality → Unique

One solution

Alternates

A few solutions

Quasi-unique

Differences are not meaningful

1D-Josephson Junction Problem

current direction

grain boundary

$I_c(B)$

$J_c(x)$

x

magnetic field
B

I_c (

Magnetic Field (B)

$$I_c(B) = |\int exp(2\pi ixB)J_c(x)dx|$$
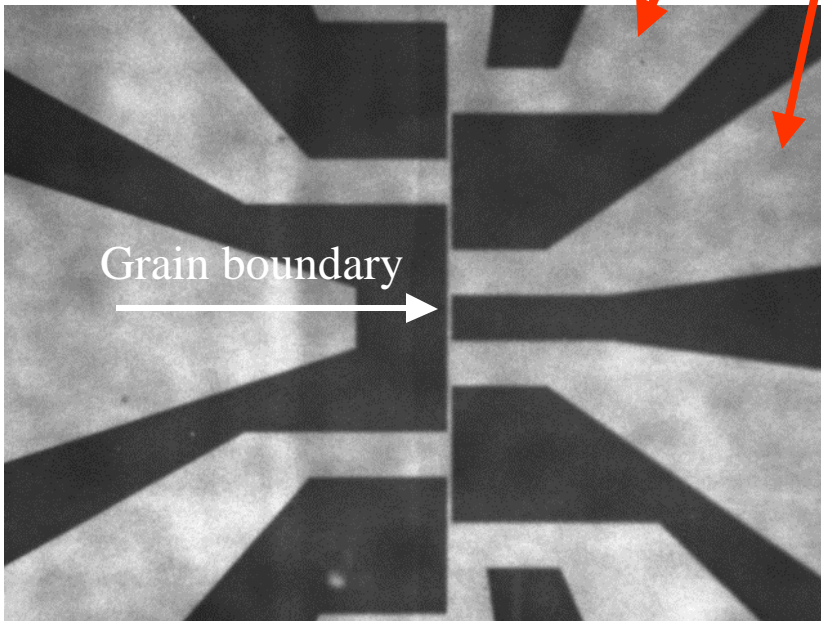
Physica C <u>315</u>, 145 (1999) ; Journal of Applied Physics 87, 2454 (2000) ;
Interface Science 8, 231 (2000); PRB submitted

[010]

[100]

[010]

[100]

5 nm

grain boundary

YBCO film

bicrystal substrate

[001]
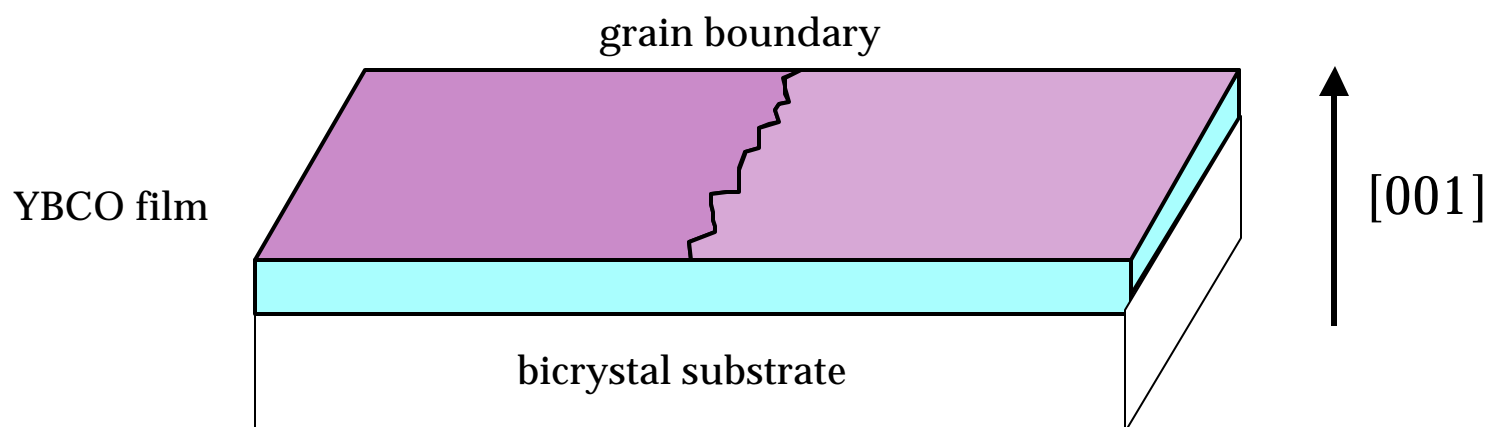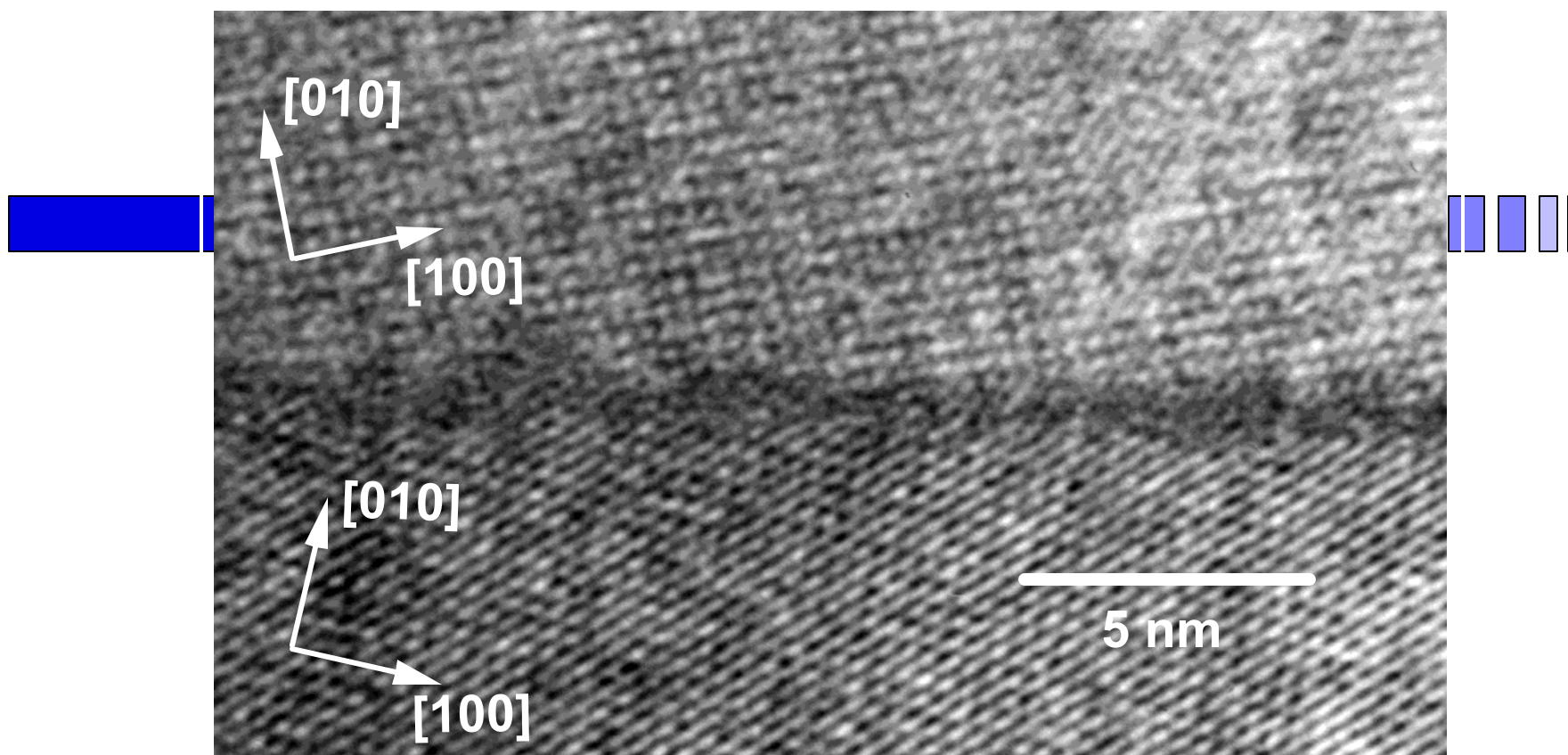
Generate a compact support

Electrical Contacts

Grain boundary

10 μm

Location of grain boundary
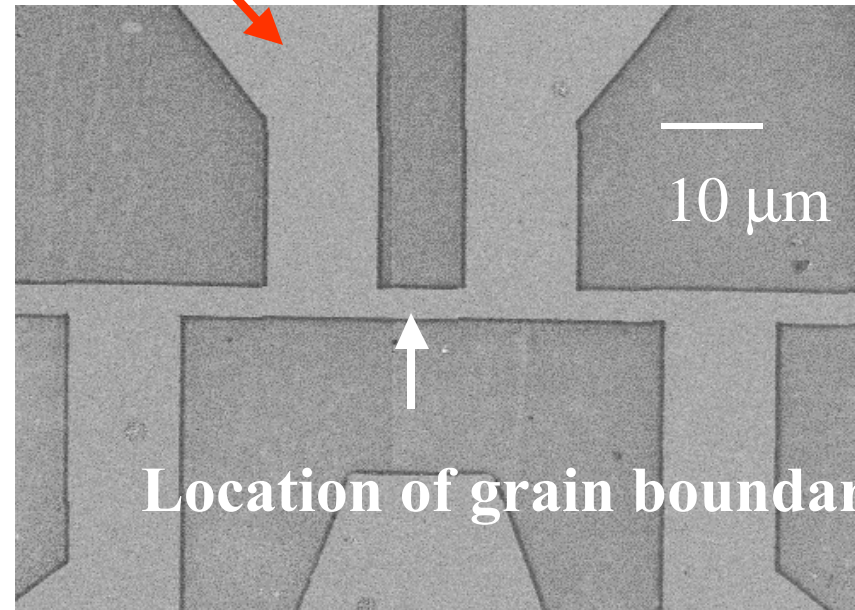
Create a 1-D finite object by micropatterning

# Constraints on Real-Space Form

- For simple (low-angle) boundaries
  - Boundary is finite
  - Current is positive
  - Current is less than a known maximum (weak)
- For 45 degree boundaries
  - Boundary is finite
  - Current may be positive or negative

Method

- Standard "HIO", i.e. successive orthogonal projections
- FOM = L1 or L2 mean (does not seem to matter *here*)
- Genetic search to find feasible set of solutions
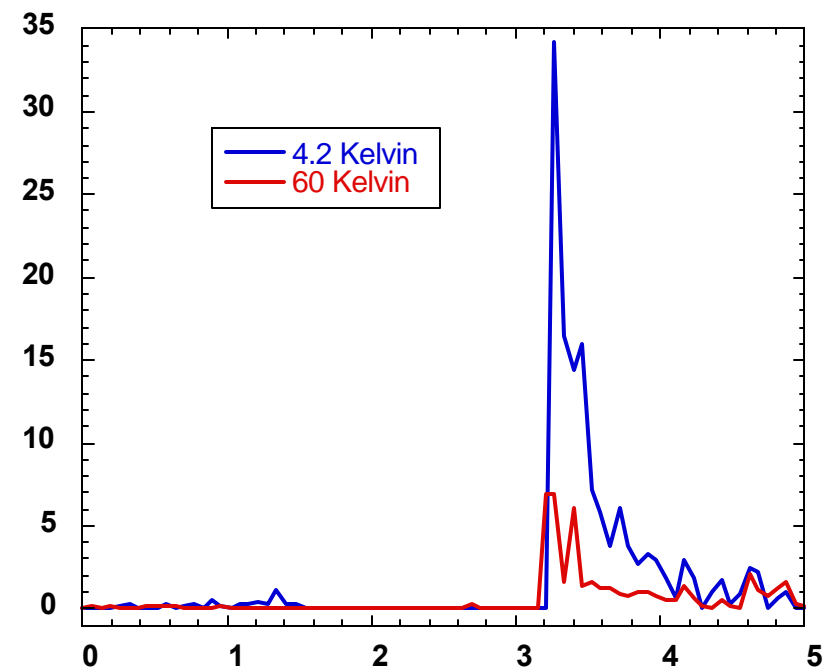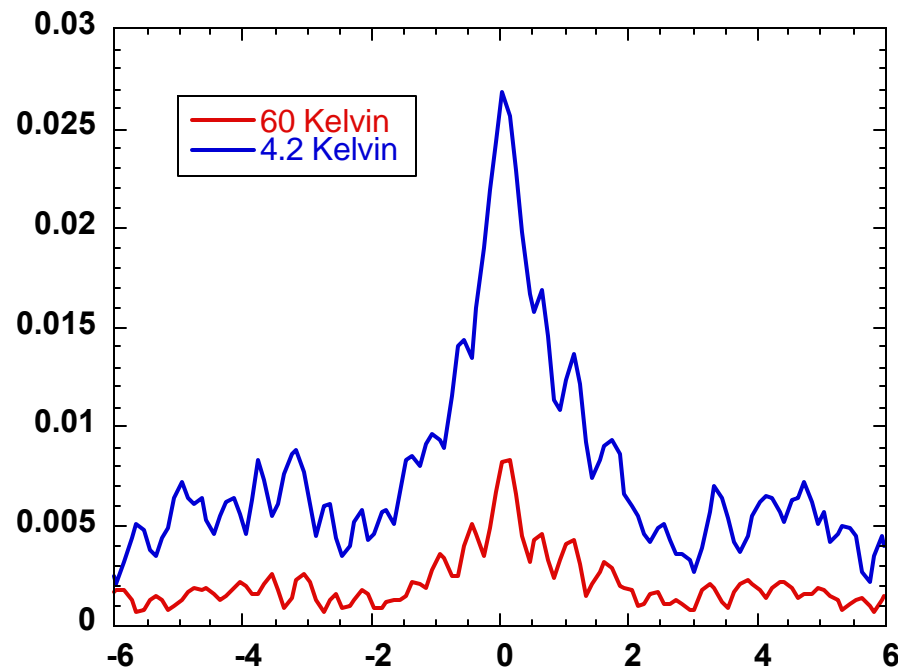 - For M initial trials, best N form the feasible set

Solutions are quasi-unique
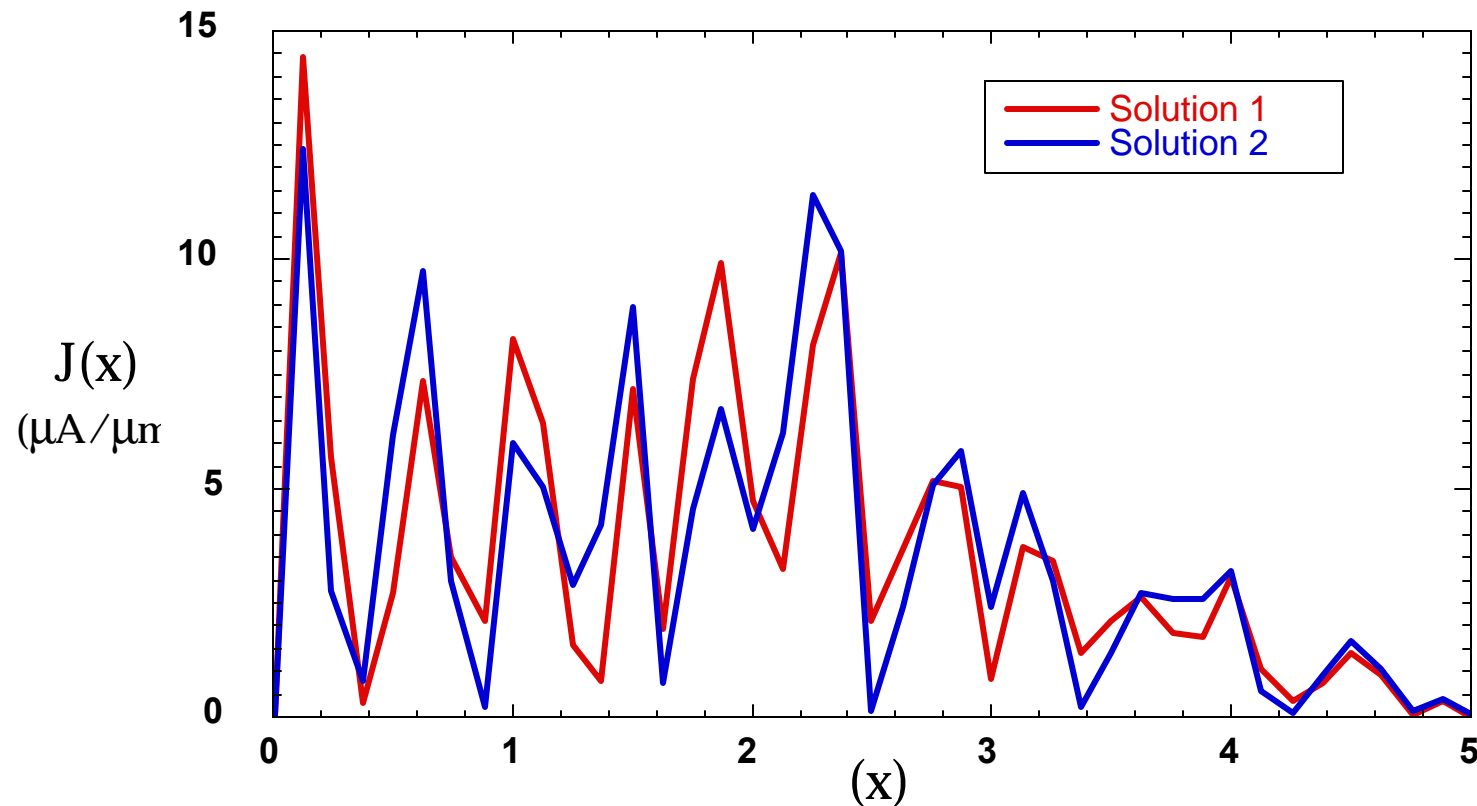
Experimental Data: J(x) > 0

24 ° YBCO Bicrystal, 5 μm Wide Boundary
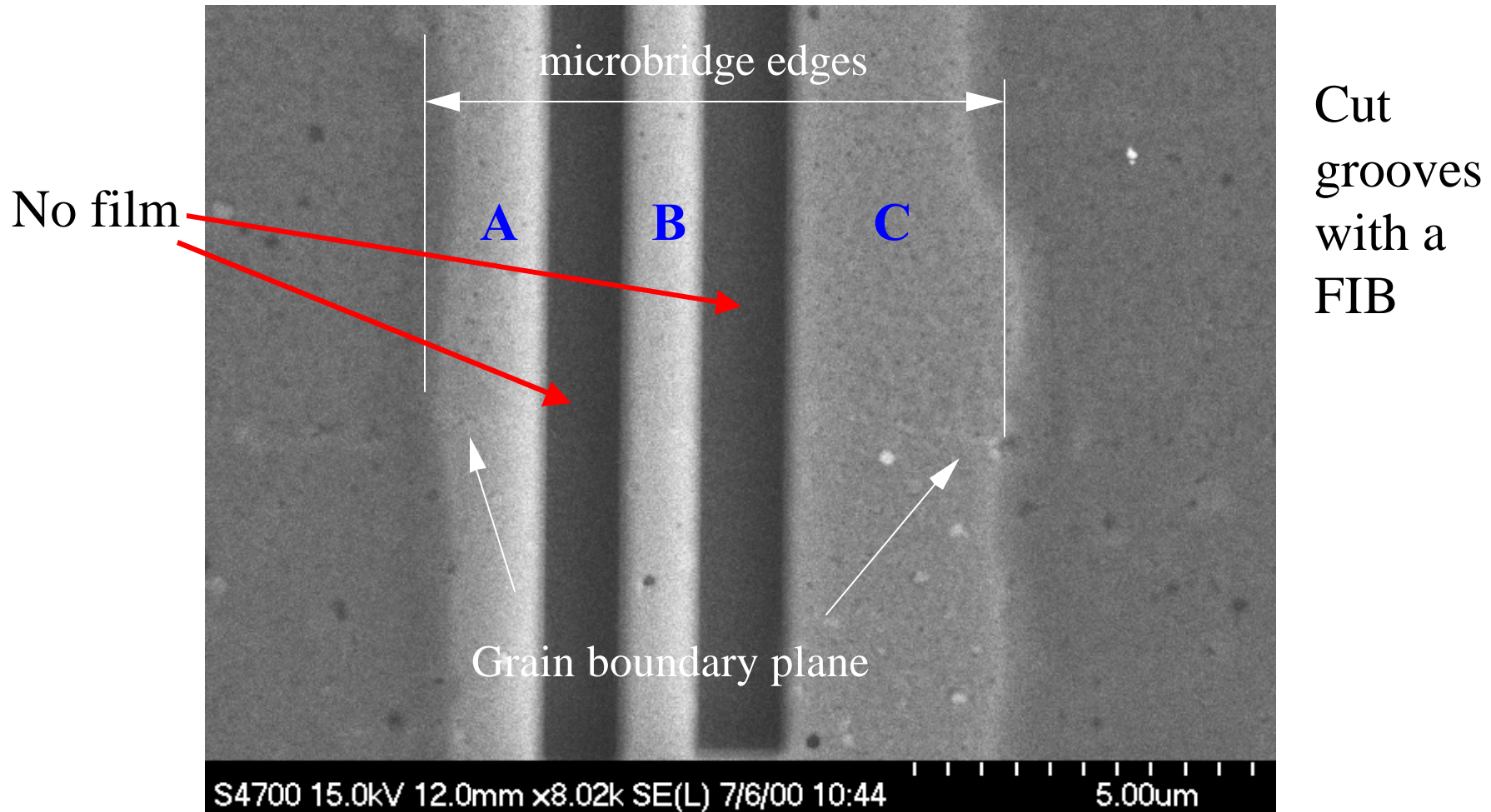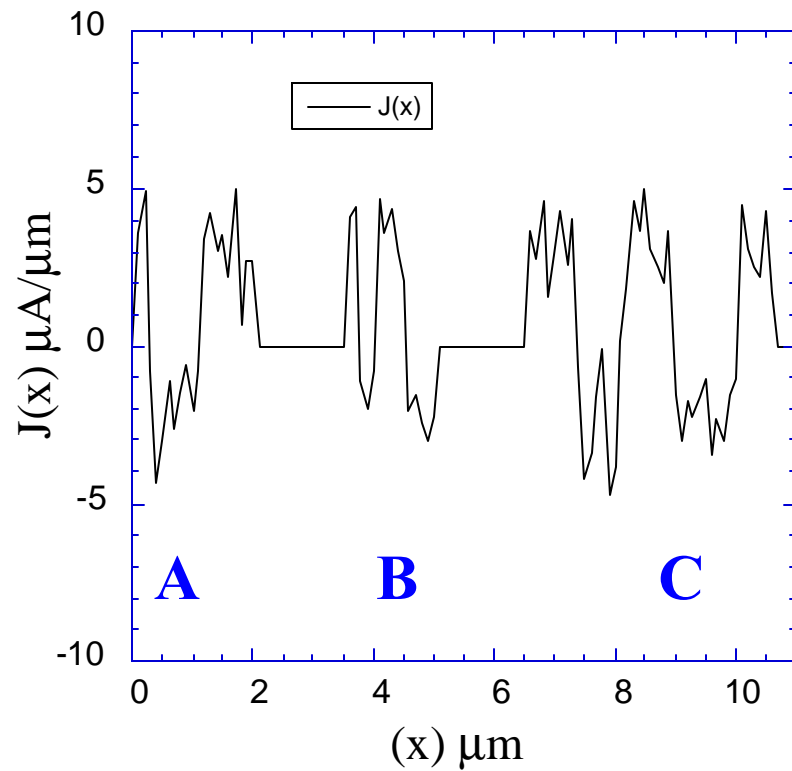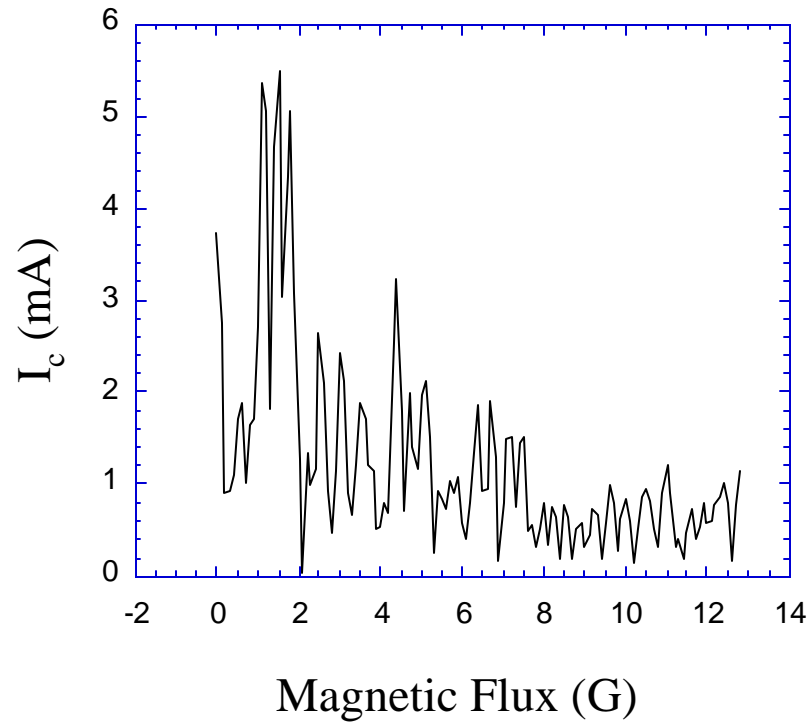
Verification via changing Temperature
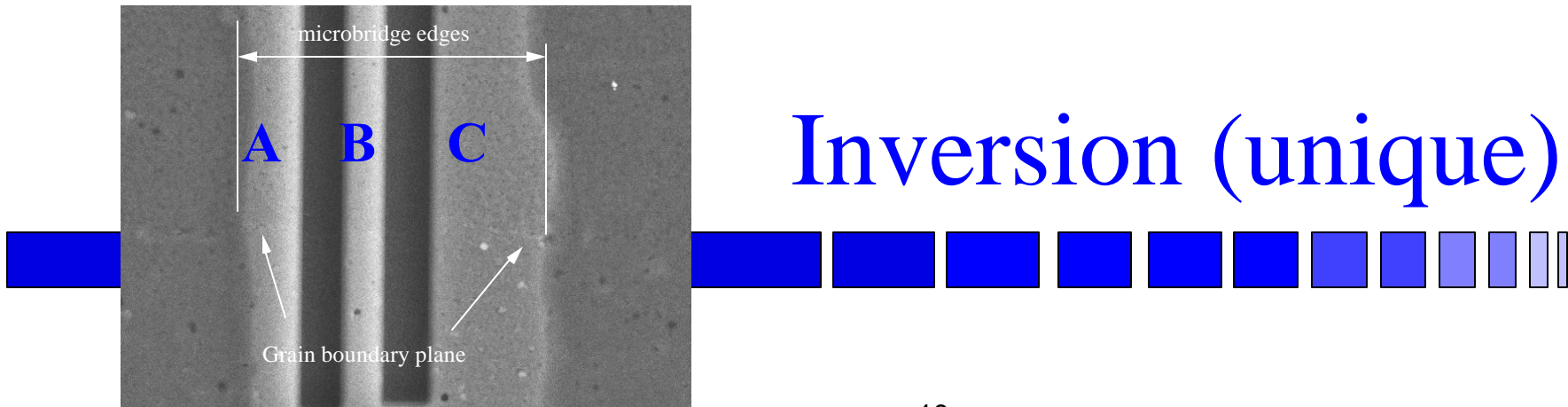

Measured Data

Inversion

Experimental Data: 5μm boundary

45° boundary: $-J_c<J(x) < J_c$
Use a discontinuous support

microbridge edges

A   B   C

No film

Cut grooves with a FIB

Grain boundary plane

S4700 15.0kV 12.0mm ×8.02k SE(L) 7/6/00 10:44   5.00um

Inversion (unique)

microbridge edges

A B C

Grain boundary plane

I_c (mA)

Magnetic Flux (G)

J(x)

$J(x)$ μA/μm

A **B** **C**

(x) μm
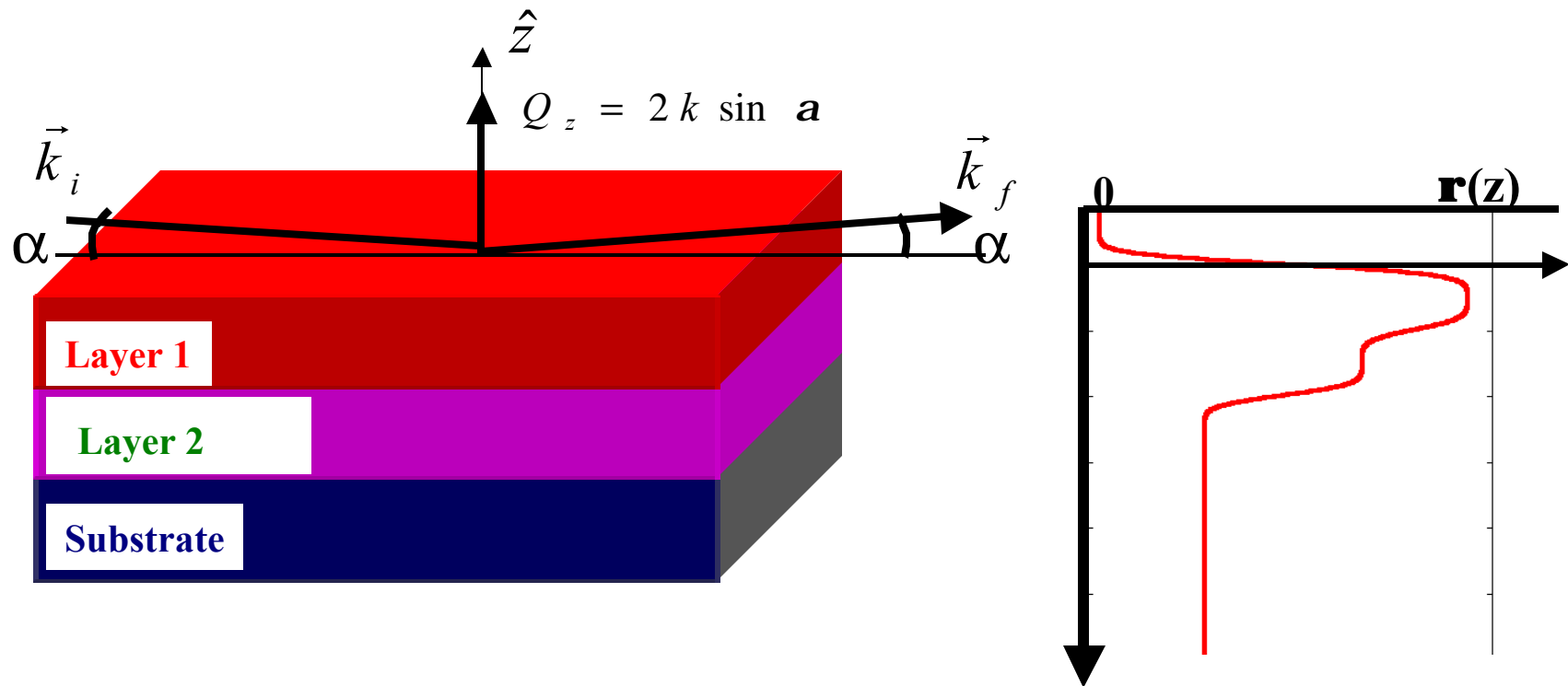
N.B.: support is smaller than that which is known to be unique
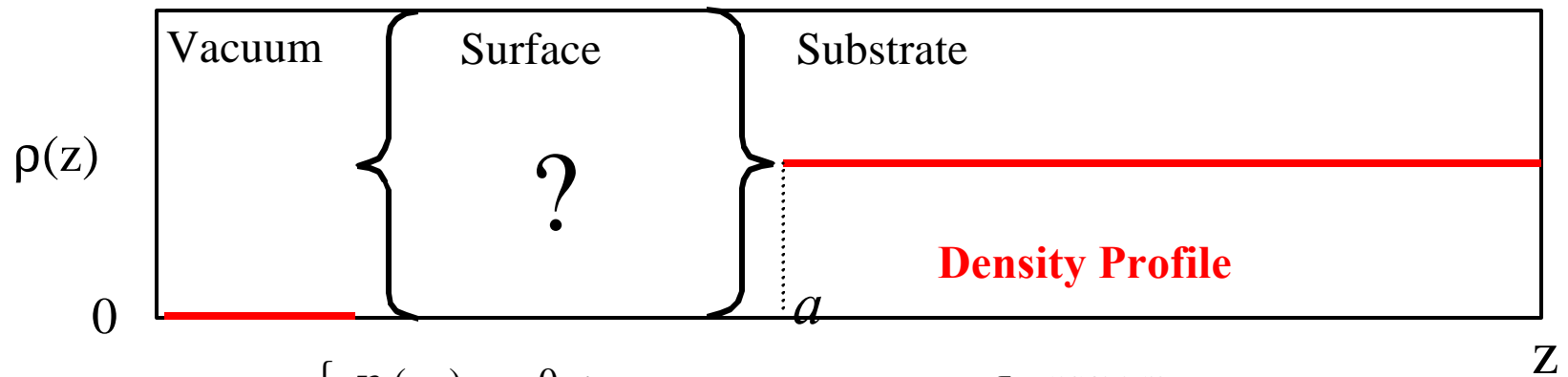
1D- X-ray Reflectivity Problem

Measurement: $I(Q) = \left| \int \mathbf{r}'(z) \exp(iQz)\,dz \right|^2$

(Kinematical Approximation)

Bengu, Salud & Marks, PRB, in press
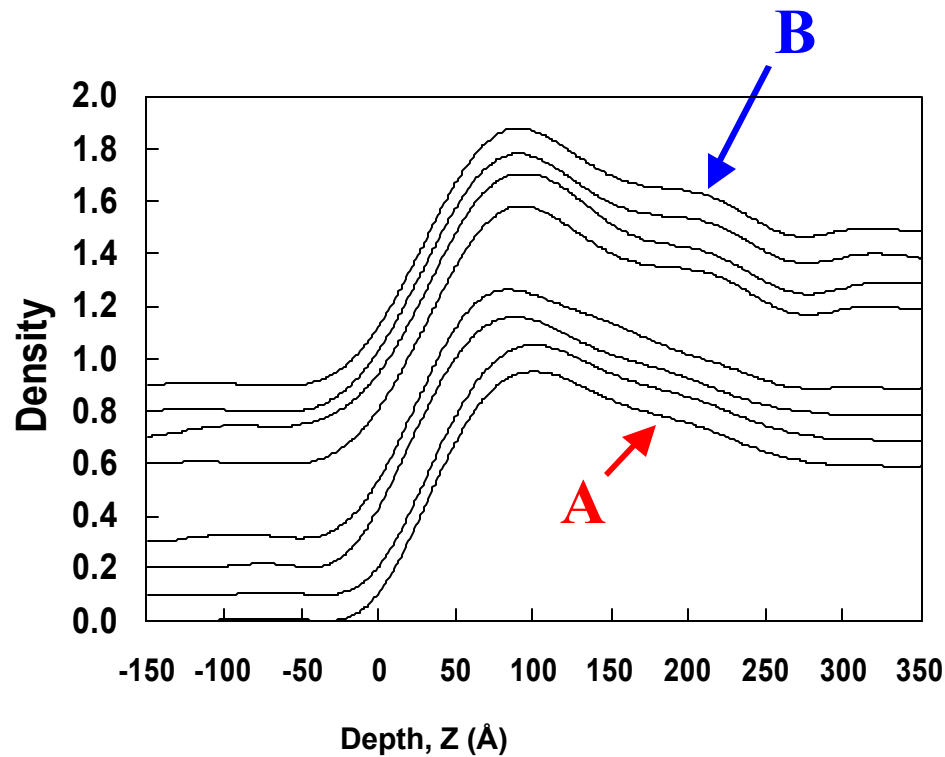
Compact Support for dρ(z)/dz

$$
S_1 = \begin{cases}
\boldsymbol{r}(z) = 0 : & z \in vacuum \\
\boldsymbol{r}(z) : & 0 \leq \boldsymbol{r}(z) \leq D_{max} \\
(1 - \boldsymbol{d}_2)\,\boldsymbol{r}(z) : & \boldsymbol{r}(z) < 0 \\
D_{max} - (\boldsymbol{d}_2 - 1)\,\boldsymbol{r}(z) : & \boldsymbol{r}(z) > D_{max} \\
\boldsymbol{r}(z) = D_{sub} : & z \geq a
\end{cases}
$$

$$
S_2 = \{\, \boldsymbol{r}(z) : \left| FT\{\boldsymbol{r}'(z)\} \right|^2 = I_0(Q) \,\}
$$

Quasi-Unique Solutions

Real space (y axis offset)

Model Data
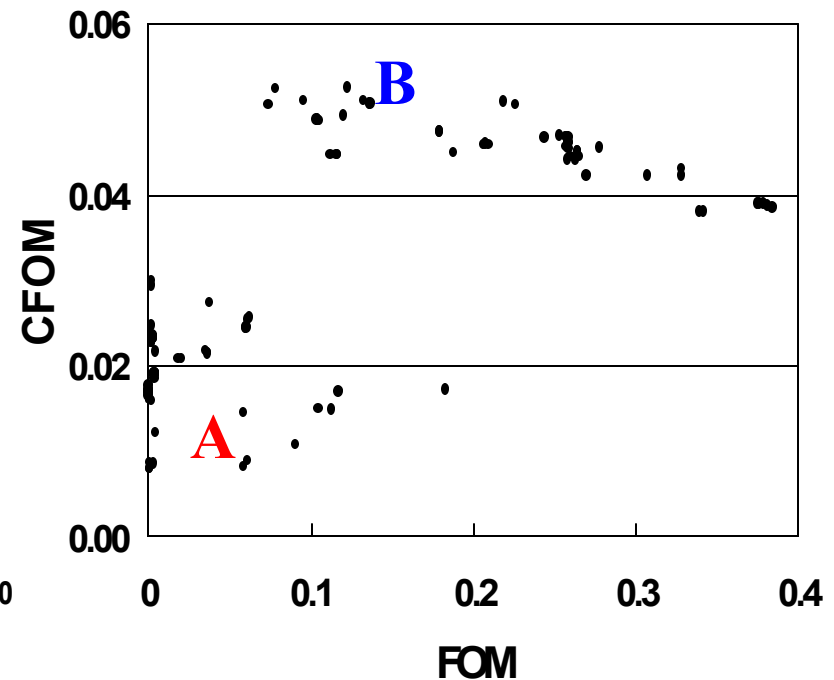
FOM versus Original (CFOM)

Experimental Data
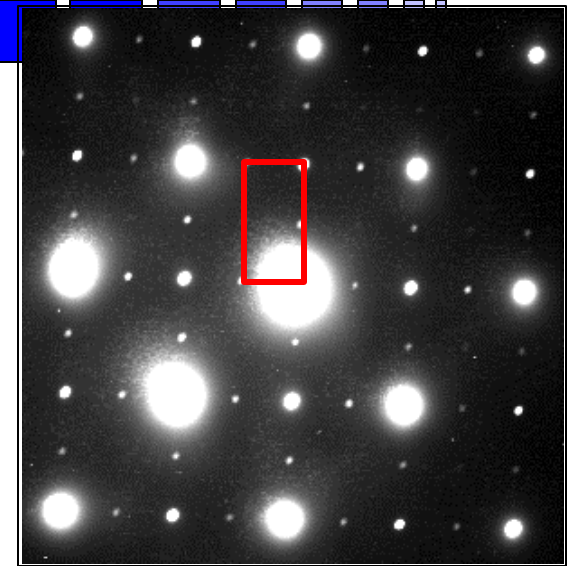

PFA on water + KOH solution, pH=11.2

19.07±0.3 Å
(19.0 Å)*
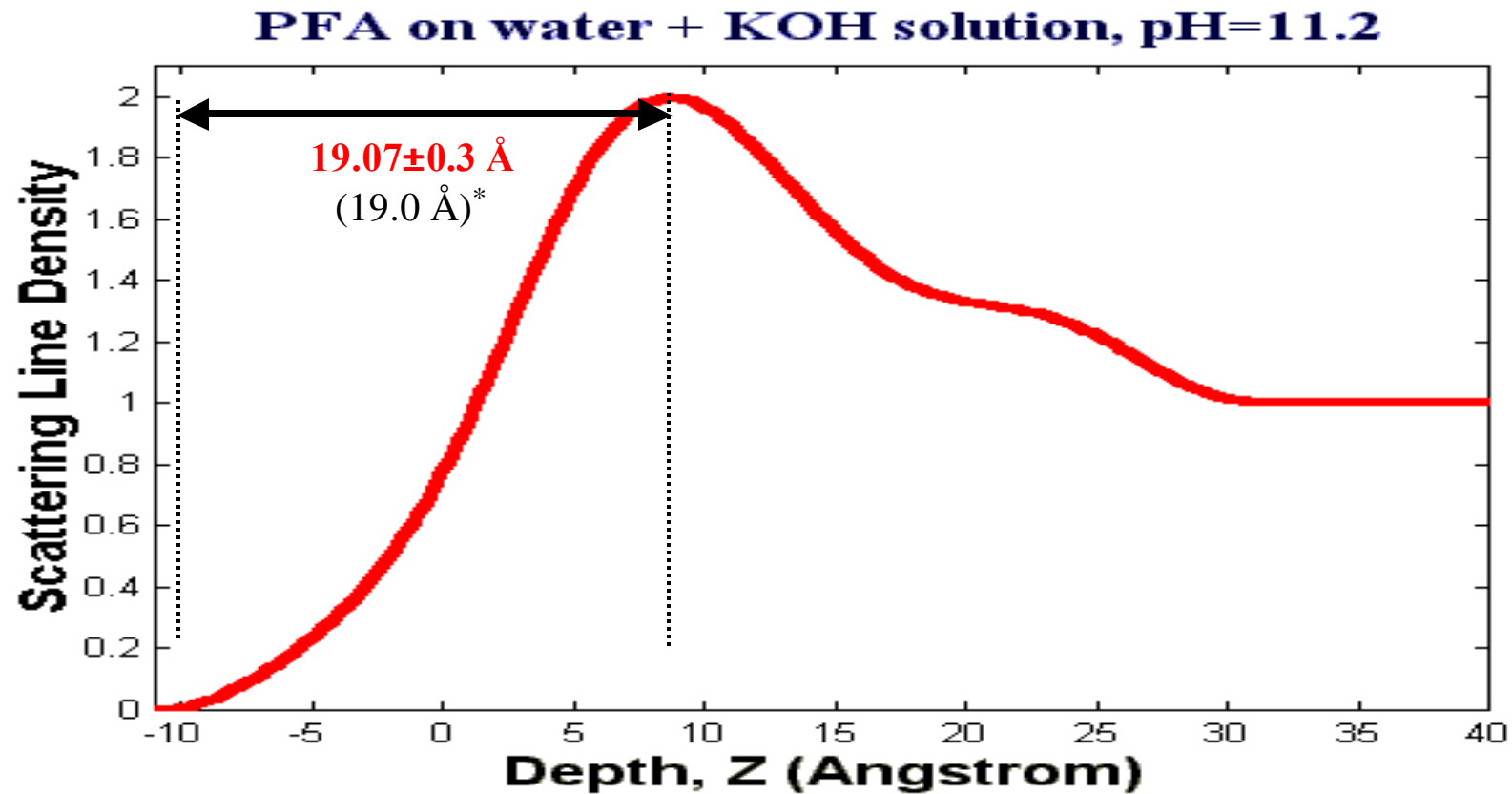
* J.S. Pedersen (1992), J. Appl. Cryst., **25**, 129.

1D/3D-Surface Problem

- Incomplete set of measurements
  - **20-30%** of total
- Atomistic constraints
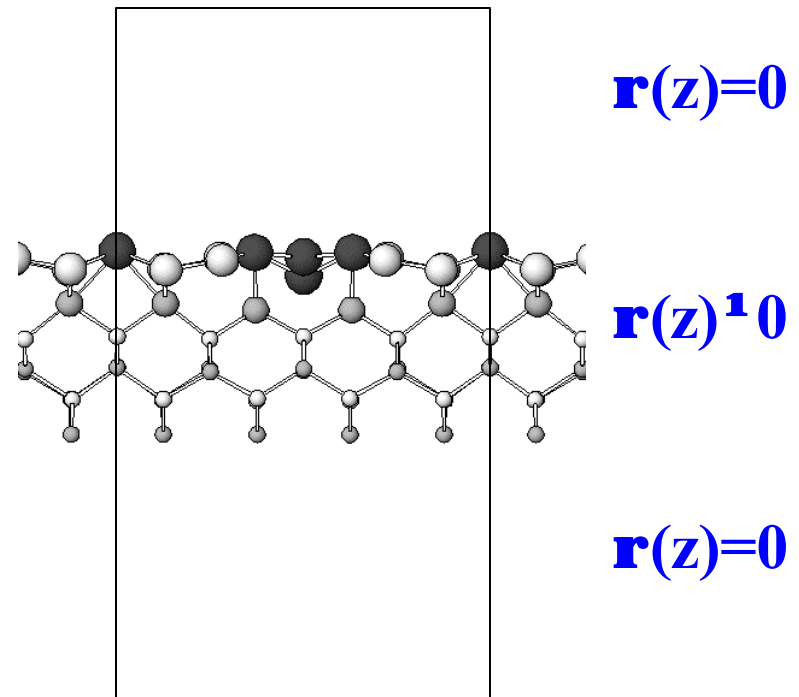- Periodic in x,y; compact support constraint along z

SrTiO$_3$ (001) 2x1

Basics: Surface Reviews and Letters <u>5</u>, 1087 (1998) ;
Acta Crystallographica <u>A55</u>, 601 (1999); Physical
Review B <u>60</u>, 2771 (1999)

3D-Support Constraint

- Displacements decay as $(\alpha+z)\exp(-qz)$ into bulk[1]
- Consider only non-bulk spots
- Real space constraint
  - $\rho(z)=0$ away from surface
- Convex constraint

$\mathbf{r}(z)=0$

$\mathbf{r}(z)\neq0$

$\mathbf{r}(z)=0$

[1]Biharmonic expansion of strain field, Surface Science <u>294</u>, 324 (1993)

Why we don't need all the data

- The constraints, e.g. support & atomistic, generate both amplitude & phase estimates.
- The amplitudes and phases of the unmeasured points must also be consistent with the constraints.
- Hence it is often (not always) possible to recover to a good approximation the "missing cone" values

Unmeasured Reflections

Recovery of Unmeasured Reflections
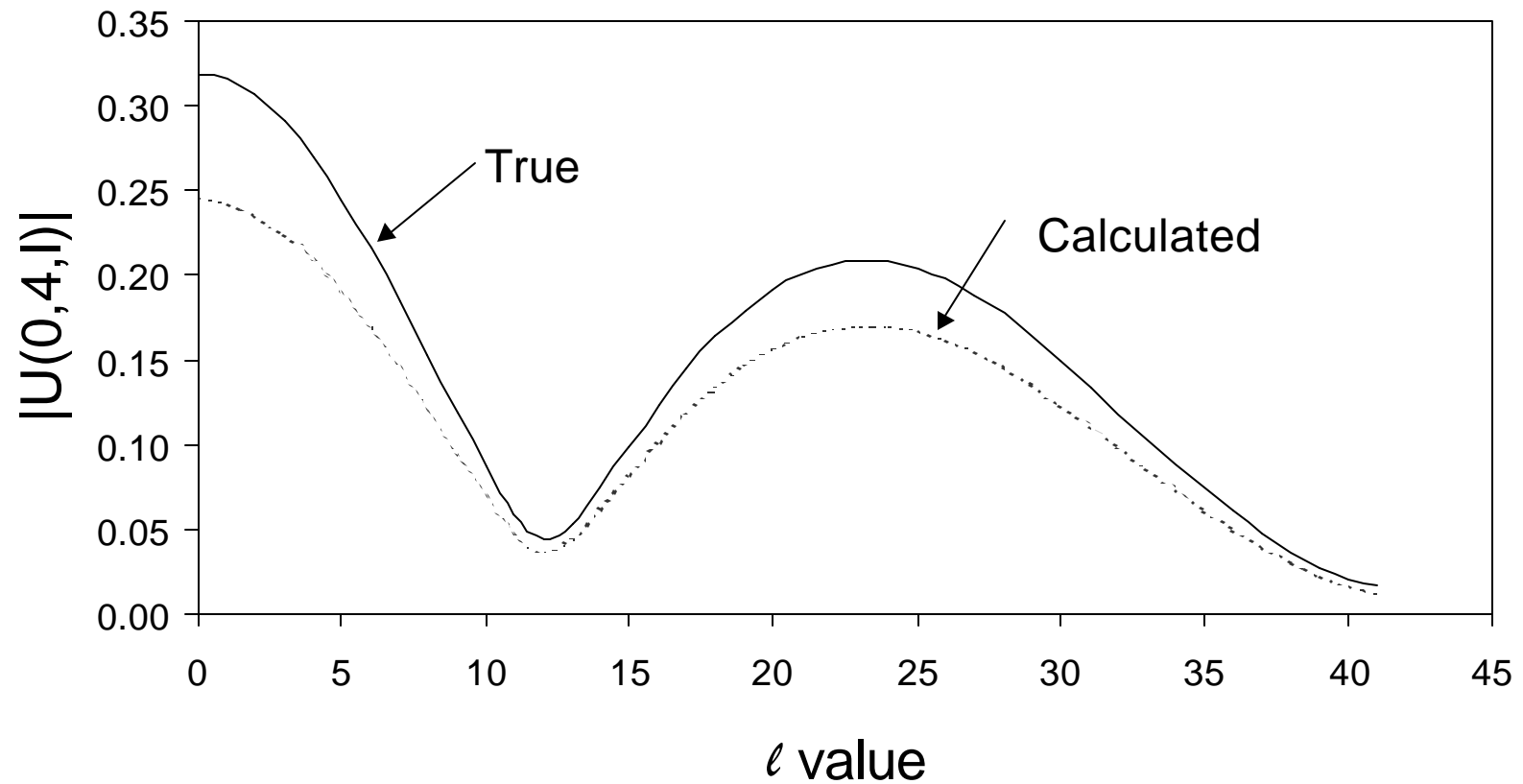
Implimentation

- 300-1000 known moduli typically
- Two weighted parallel "atomistic" operators
- Over-relaxation & extrapolation
- Some "Statistical" constraints (set large $U(k)$ first)
- L1 FOM (much better than L2)
- Genetic Algorithm global search – 1000 to 10000 initial phase sets (1-4 hrs on an HP workstation)
- About 20 3D FFT's per starting point (10 iterations)
- Projection onto known atomic positions (as they become available)

Overall methodology

- Solve simplest problem with no prior information first

- Add additional constraints as analysis progresses
  - Pruning of unrealistic solutions
  - Acceptance of "correct" elements (e.g. atoms)
- Tends (hopefully) to a unique solution

Many subtle points

- Consider the FOM = $| F_{true} - F_{est}|^2$
- Error Gaussian
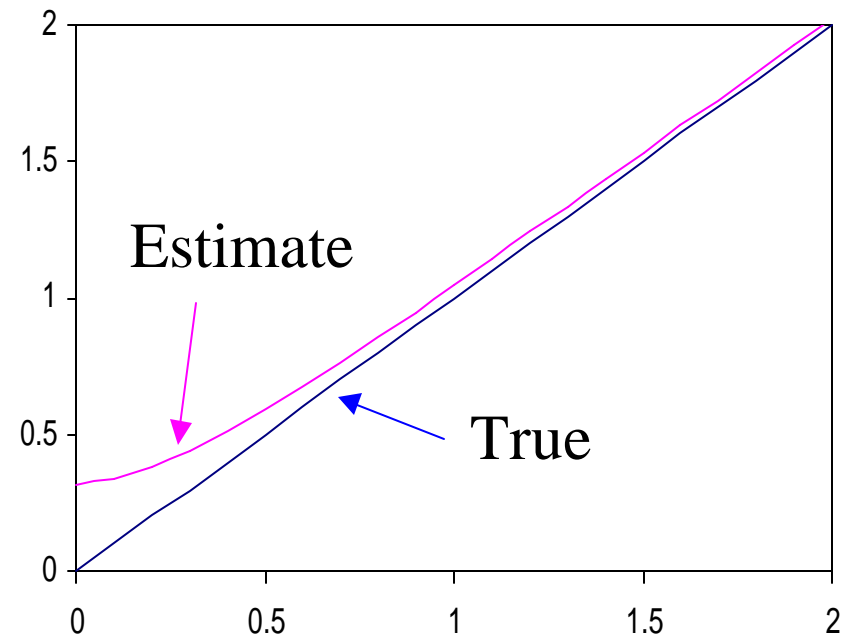 - $F_{est} = F_{true} + noise$
- F_{true} large
  - $<F_{est}> \sim <F_{true}>$
- F_{true} small
  - $<F_{est}> \sim noise$
    $>> F_{true}$
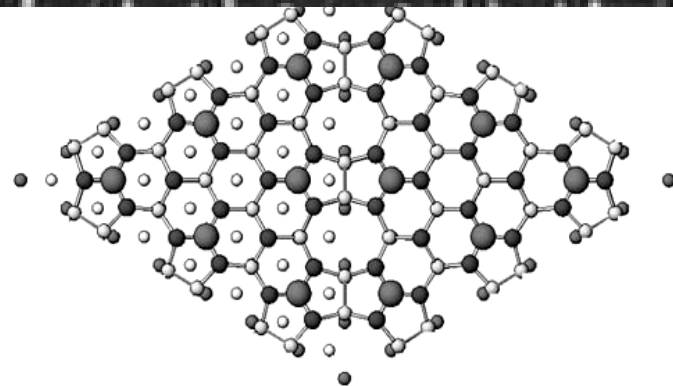 (similar to SIM weights)

Role of "background" compensation

- Si (7x7) in p3m1 without compensation

Experimental Data

Note: in p6mm compensation is not needed
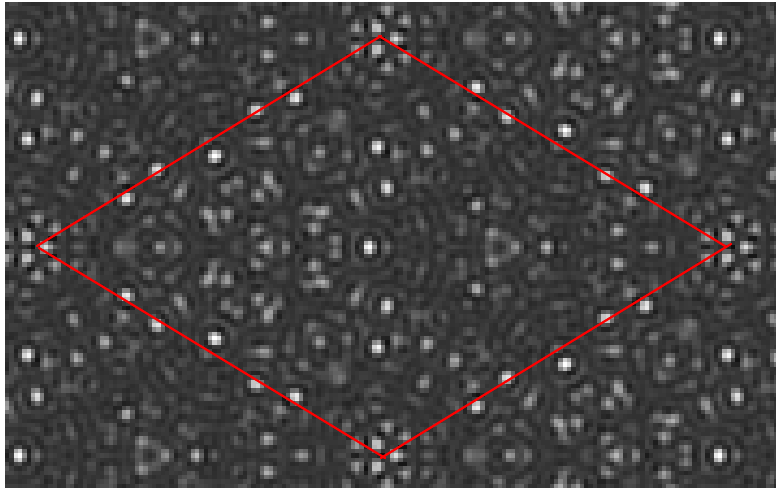
- Si (7x7) in p3m1 with compensation
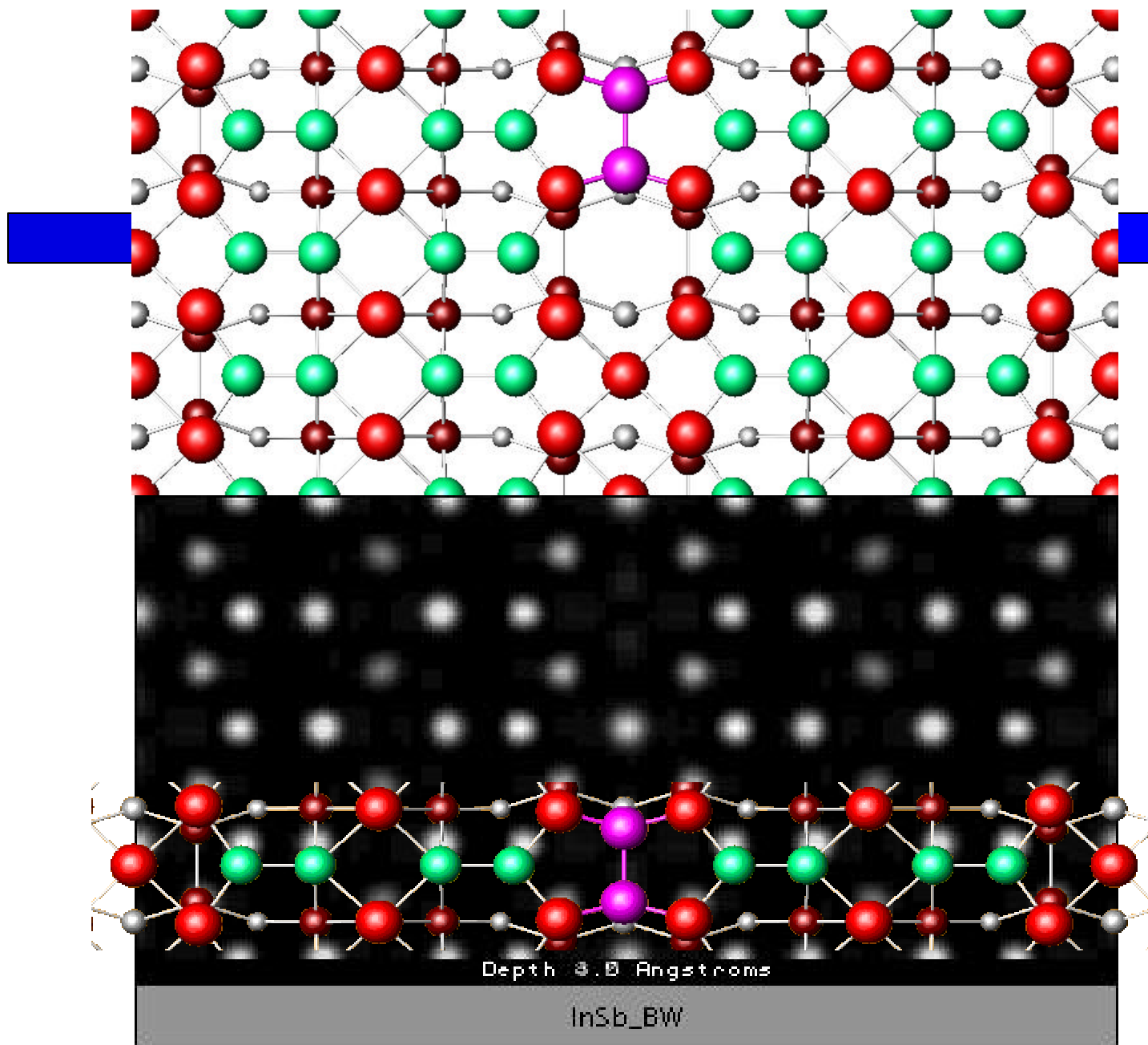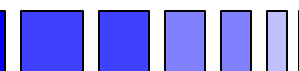
NiO (111) 2x2 (p3m1)

Experimental Data: Solution is quasi-unique in 3D (two branches in 2D)

NiO 2x2 Depth 0.8 Angstroms
NiO_BW
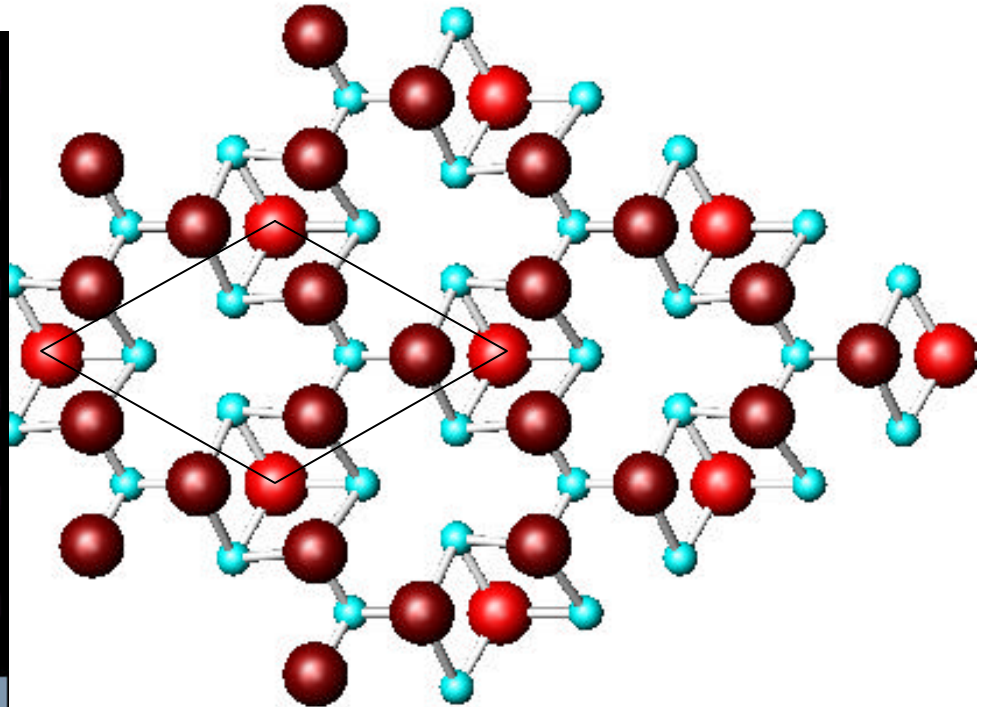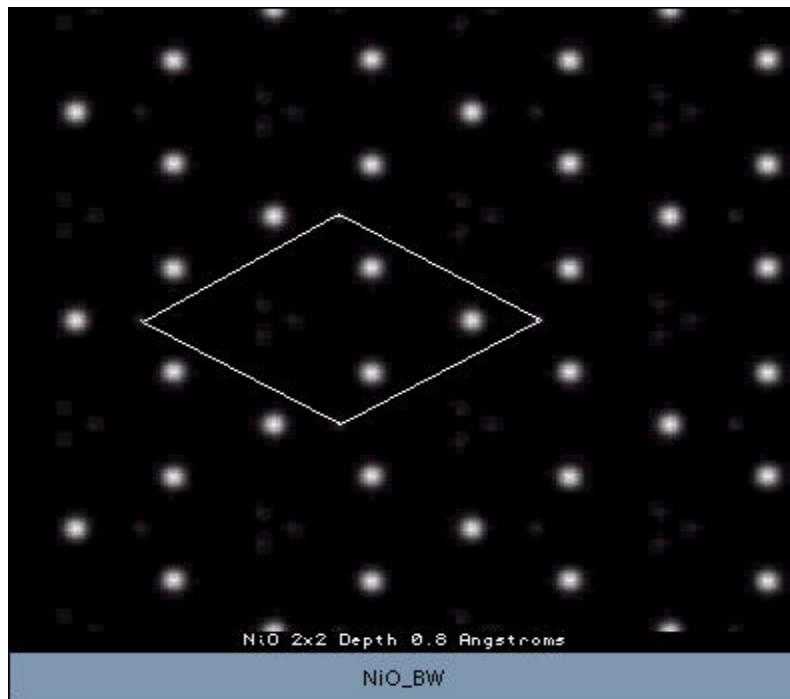
Surface Science <u>470</u>, 1-2, 1 (2000)

InSb c8x2

Experimental Data

Quasi-unique in 2D & 3D

Physical Review Letters 86, 3586 (2001)

Conclusions

- We don't need a formally exact recovery of the phases, only an approximate one

- We can generalize to include atomistic and other constraints

- Many 1D problems are quasi-unique

- Many 3D crystallographic problems are quasi-unique